[redacted]

RADIOACTIVE ISOTOPES OF IODINE

B. G. Dzantiyev and M. B. Neyman

Uspekhi Fizicheskikh Nauk, Vol XXXV, No 2, June 1948

[redacted]

3 August 1950

SECRET

USPEKHI FIZICHESKIKH NAUK
Vol XXXV, No 2, June 1948

RADIOACTIVE ISOTOPES OF IODINE

B. G. Dzantiyev
M. B. Neyman

INTRODUCTION

The use of artificially produced radioelements in chemistry, physics and biology necessitates a knowledge of their physical characteristics.

Depending upon the nature of the problems to be solved by the application of radioactive isotopes, chemical, physiological, or physical properties must be studied. In many cases it is necessary that an element of given chemical behavior and physiological action should at the same time possess definite physical properties; e.g., have a definite half-life, emit beta-particles and gamma-quanta of definite energy.

In many cases these requirements can be satisfied by a given chemical element in the form of a radioactive isotope. To a considerable extent this refers to both radioiodine, which has been widely used for some time as an indicator in various processes and as a therapeutic agent. By means of this element, Soviet research workers have solved many problems in the field of chemical kinetics (reaction mechanism), the chemistry of complex compounds, the structure of solutions, the mobility of atoms in crystal lattices, etc. In most of this work, use was made of the iodine isotope with a half-life of 25 minutes.

At present some 14 radioactive isotopes of iodine are known which have half-lives ranging from several seconds to several months, as well as various types of energy radiation are known. A rational use of these isotopes, taking into account their physical characteristics, permits one to treat a considerable number of problems.

SECRET

Some isotopes of iodine which are of great interest have been discovered very recently.

Preparation and Isolation of ISOTOPES OF IODINE

The mass-numbers of iodine isotopes lie in the interval M 124----137, and only one of these isotopes is stable [1].

A considerable number of these isotopes are separated from the fission products of neutron-bombarded uranium and thorium. Some radioactive isotopes are obtained by bombarding the stable isotopes of I, Te, Sb, and Cs with neutrons, protons, deuterons, and alpha-particles. It is obviously possible to obtain iodine also by the neutron bombardment of xenon involving the reaction Xe_{54} (n, p) I_{53}; but, as far as we know, such a reaction has not been carried out. The essential advantage of radioactive iodine over many other artificially produced radioelements is its relative ease of separation from the bombarded target and from secondary nuclear reaction radioactive products. The elementary state [2], the chemical method generally employed in separating radioiodine from irradiated targets consists of oxidizing iodine to its elementary state and separating it from a solution of the original target by utilizing either the high vapor tension of elementary iodine or its solubility in organic solvents. The carrier is added to the exposed target, generally either in the form of KI or of an equivalent mixture, KI + KIO_3. In the former case, to oxidize the iodine, "soft" oxidizing agents of the type KNO_2 or $Fe_2(SO_4)_3$ are used which do not affect other halogen. This is particularly important in separating iodine from the fission products of heavy nuclei. In the latter case, elementary iodine is separated through acidification of the solution according to the reaction:

$$5KI + KIO_2 + 3H_2SO_4 = 3I_2 + 3K_2SO_4 + 3H_2O.$$

SECRET

SECRET

There is good reason to assume that this method achieves the most complete quantitative separation of iodine, since in this event in the process of transition to elementary state all atoms radioactive must be involved, no matter what the valency state may be as a result of nuclear reaction.

In cases where there is some likelihood of a joint oxidation of bromine, a certain amount of KBr besides KI must be added to the solution to lessen the probability of separating active bromine.

If stable iodine serves as the target, in accordance with the method of Szilard and Chalmers further developed by many other authors, the concentration of a weighable quantity of a radioactive isotope and its separation from stable iodine is produced by utilizing the change in valency and chemical behavior of the atom obtained as a result of nuclear reaction. Separated by one method or another, the elementary iodine containing active isotopes is separated from the solution target either by agitating it with corresponding organic solvents (CCl_4 or $CHCl_3$) or by distilling it from a boiling solution heated by steam. As a result of these operations it is possible to avoid the necessity, usual in the majority of other cases, of precipitating the required element directly in a target solution. Thus, in this case we eliminate the danger of simultaneous precipitation and absorption of other radioactive elements, which may have formed jointly with the iodine during nuclear reaction. In some cases, in addition to the above-mentioned chemical methods, electrolytic separation of active iodine is employed directly during irradiation.

ISOTOPES OF IODINE AND THEIR PHYSICAL CHARACTERISTICS

In studying separate isotopes of iodine, we shall take up the following problems: (1) nuclear reactions; (2) the type of radioactive decay reaction; (3) the mass number; (4) the mass number corresponding to the given activity; and (5) the beta-spectrum, the gamma-rays, and decay.

SECRET

I 124, T = 4 Days

The isotope with A equals 124 has the lowest mass number of all known isotopes of iodine. It was obtained by Livingood and Seaborg by bombarding antimony with beta-particles with energy E equals 16 Mev. In measuring the activity of iodine separated from antimony exposed to beta-particles, the decay curve shown in Figure 1 was obtained. This curve can be represented as the result of superposing two curves of periods T equals 4.0 equals 0.3 days and T equals 13.0 equals 0.3 days. The existence of two periods agrees with the fact that antimony has two stable isotopes, Sb 121 (56 percent) and Sb 123 (44%), from which two isotopes of iodine, I 124 and I 126, are formed by the action of alpha-particles according to the reaction Sb 51 (alpha, n) I 53. Since, as the result of other nuclear reactions, it has been shown that a 13-day period must belong to I 126, then activity with T equals 4 days must correspond to I 124. This isotope, according to Livingood and Seaborg, was obtained according to the reaction Sb 121 51 (alpha, n) I 124 53 with the discharge of 1 active atom per 2.107 bombarding alpha-particles of energy E equals 16 Mev. I 124 emits positrons and forms a stable tellurium: I 124 → Te 124 + beta-plus.

Figure 1. Decomposition curves of I 124 and I 126. 1. Decomposition of radioactive isotopes of iodine formed according to the reaction Sb (alpha, n) I. 2. Decomposition of I 126 formed during the irradiation of iodine by fast neutrons.

The spectrum of positions emitted by I 124 and the presence of gamma-rays during beta-decomposition were not studied.

Reference to the bibliography shows that an isotope of iodine was also obtained by Dunbridge by bombarding tellurium with protons according to a Te (p,n) I reaction.

SECRET

It is curious that, in bombarding tellurium with deuterons of energies equal to 8 Mev and E equal to 11.5 Mev, the emergence of a positron of half-life 40 days was not detected [8, 10, 11], although it was possible to expect its formation according to the reactions (d,n) and (d,2n).

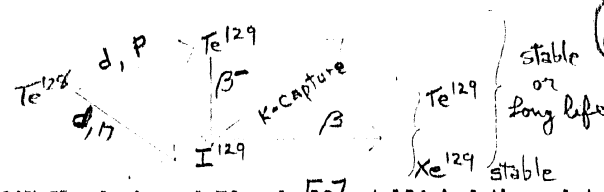
SECRET

I 125, T = 56 days.

Until recently there was no radioactive isotope of iodine to which a mass number M equal to 125 could be assigned. There is no isotope I 125 in Seaborg's isotope table. In 1946 Reid and Keston [12] announced that in bombarding tellurium for 40 hours with deuterons of energy 14.5 Mev they had obtained a long-lived activity which did not die out for 6 months. This activity is due to the iodine during precipitation of AgI and extraction with CCl_4; when introduced into mice, it concentrated in the thyroid gland. The half-life of this activity, which obviously pertains to iodine, proved equal to T equal to 56 days.

In studying the radiation emitted by these radioactive isotopes, by means of absorption in aluminum and copper and silver foil, a characteristic X-ray radiation of energy E equal to 27.0 Kev was detected, which corresponds to the K-alpha line of Te. Gamma-rays of greater energy were not discovered. Hence the authors came to the conclusion that the radioactive transmutation of the new iodine isotope chiefly resulted in K-capture with transition to Te. Simultaneously they proposed on insufficient data that the 56-day period belonged to I^{129} and suggested a complicated scheme of transmutations including the unique case consisting of successive beta-decay and K-capture (see diagram below).

(X) on Page 15



In 1947 Glendenin and Edwards [13] established the existence of an iodine isotope of half-life T equal to 56 days but ascribed this time to I 125. In this case the 56-day activity was isolated from tellurium deuteron-bombarded

SECRET

about a year earlier. The chemical identity of this activity with iodine was demonstrated by a series of successive operations of oxidizing iodine with sodium nitrate, extracting the CCl_4 from it, and reducing with a solution of sodium sulfide; whereupon it seemed evident that the 56-day activity in all operations is due to the iodine. As a result of three cycles of such operations, the initial activity remained unchanged within the range of experimental error.

In studying the absorption of the radiation emitted by 56-day iodine in Ag, Cd, In, Sn, Al, and polystyrene, only K-ray radiation of energy E equal 27.5 Kev and E equals 3.8 Kev was discovered, this corresponds with the K-alpha and L-alpha lines of tellurium. No gamma-ray of energy E greater than 30 Kev and no particles were discovered in the radiation of 56-day iodine. Hence, it is obviously possible to consider proved that a radioactive iodine isotope of half-life T equals 56 days exists and that its radioactive decay results in obvious K-capture.

With regard to the mass number of 56-day iodine, this half-life should probably be ascribed to I 125.

Reid and Keston's proposition must be considered erroneous on the basis of the general ideas [13] as to the probable character of the radioactive transmutation of a nucleus with Z equals 53 and M equals 129 and on the basis of experimental data, which will be treated later, and from which it is possible to conclude that a different half-life must be attributed to I 129.

On the other hand, it is possible for I 125 to be formed by bombarding with deuterons the stable isotopes of Te 124 (4.5%) and Te 125 (6%) according to the reactions $Te\ {}^{124}_{51}\ (d,n)\ I\ {}^{125}_{53}$ and $Te\ {}^{125}_{52}\ (d,2n)\ I\ {}^{125}_{53}$. Radioactive decay through K-capture by I 125 is not unexpected (compare the stable I 124 series undergoing the same process of nuclear transmutation p, n) and leads directly to stable Te 125.

Here the artificial assumptions required by Reid and Keston are not needed.

SECRET

All these facts indicate conclusively enough, though somewhat indirectly, that the 56-day half-life must belong to I 125. However, there are as yet no direct proofs of the identity of the iodine isotope of half-life T equals 56 days with I 125.

I 126. T = 13 days

In bombarding stable iodine with fast neutrons, Tape and Cork [14] discovered a radioactive iodine isotope of half-life 13 days to which they attributed the mass number M equals 126. By observations in a Wilson chamber it was established that 13-day iodine emitted electrons during radioactive decay.

The existence of an electron-emitting iodine of half-life 13 days was substantiated by Livingood and Seaborg [8, 15], who obtained it in exposing stable iodine to the action of fast neutrons and also separated it from antimony bombarded by alpha-particles of energy E equals 16 Mev and from tellurium activated by deuterons of energy E equals 8 Mev.

The decay of 13-day iodine separated from antimony and from stable iodine is shown in Figure 1.

As a consequence of the presence in tellurium of a great number of stable isotopes, the decay curve of iodine isolated from tellurium shows various half-lives. Analysis of the end of the integral curve, taking into account the propagative properties of various stable isotopes of tellurium, permits one to conclude that the mixture of radioactive iodine isotopes contains a component possessing a half-life of 13 days.

In Seaborg's opinion [9], this same isotope of iodine had been obtained by Dewbridge when he bombarded tellurium with protons according to the (p,n) reaction.

Formation of 13-day iodine is possible according to four independent reactions:

$$I^{127}_{53}\ (n, 2n)\ I^{126}_{53}; \qquad Sb^{123}_{51}\ (alpha, n)\ I^{126}_{53};$$

$$Te^{125}_{52}\ (d, n)\ I^{126}_{53}; \qquad Te^{126}_{52}\ (p, n)\ I^{126}_{53}.$$

SECRET

7

This is convincing proof that mass number 126 must correspond to half-life T = 13 days.

The radiation emitted by I 126 was studied by Livingood and Seaborg by noting its absorption in Al and Pb, and by Tape [16] with the aid of a Wilson chamber in a magnetic field. During absorption measurements there were discovered beta-particles which had a range R = 0.44 grams per square centimeter of Al and gamma-rays which were half-absorbed in 4.5 grams per square centimeter of Pb. Hence, the maximum energy of beta-particles, as computed by Feather's [17] formula $R = 0.543 E_m - 0.160$, equals $E_m = 1.1$ Mev. The energy of the gamma-rays is E = 0.5 Mev.

With the aid of a Wilson chamber in a magnetic field, Tape studied the beta-spectrum of radioactive iodine obtained by bombarding $NaIO_3$ with fast (Li, D) neutrons. On the basis of measurements of 1060 tracks, a simple beta-spectrum of energy $E_m = 1.20 \pm 0.03$ Mev was obtained. By extrapolating F. Curie's [18] graph, which is plotted in accordance with Fermi's theory of beta-decay and employing coordinates $\left(\frac{N}{p}\right)^{\frac{1}{2}}$ and E, one finds the maximum energy of the beta-spectrum to be E_m equals 1.22 Mev.

Thus, within the limits of accuracy for the data at hand, it is possible to represent I 126 → Xe 126 in the form of the graph of Figure 2.

Figure 2. Diagram of the decay I 126 → Xe 126.

In connection with the fact that there are indications of the presence of X-ray radiation in long-lived iodine [11], it is possible that there is some likelihood of another scheme of decay for I 126 involving K-capture with transition to Te 126.

SECRET

I 127.

From the mass-spectrographic measurements of Nier [17], it follows that I 127 is the only stable isotope of iodine. The presence in raw iodine of other stable isotopes of iodine is possible in the quantities relative to I 127:

I 123 1/50,000; I 124 1/50,000; I 125 1/50,000;

I 126 1/25,000; I 128 1/15,000; I 129 1/40,000;

I 130 1/120,000; I 131 1/250,000;

I 128. T = 25 Minutes

In the first works on obtaining artificially produced radioactive elements through the action of neutrons, Fermi, Amaldi and others [5,19], in exposing iodine to slow neutrons, discovered activity of half-life T ≈ 25 minutes due to the iodine precipitated in the form of AgI in the presence of Te and Sb. This activity was attributed to I 128 formed during radiation by neutron capture from stable iodine according to the reaction: $I\ 127_{53}(n, \gamma)\ I\ 128_{53}$. Then radioactive iodine was obtained and studied in a number of laboratories.

The study and use of I 128 was considerably facilitated by the Szilard and Chalmers discovered that a small quantity of active isotopes can be separated from the mass of inactive isotopes. The method for isolating isotopes chemically, subsequently widely used, was at first worked out only for the reaction I 127 (n, gamma) I 128 from the mass of inactive ethyliodide C_2H_5I 127.

It is interesting that this process, which forms the basis for utilization of the emission by the atom in emitting a gamma-quantum, permits a peculiar reversal. The emission which takes place as a result of radiation neutron capture can be used not only to extract an active atom from a molecule but also to introduce the active atom into the molecule of any substance. Thus, by exposing an iodine solution to slow neutrons in normal pentane, Reid [20], discovered that about 40% of the activity seemed to be connected with n-amyl iodine. This is obviously

SECRET

the process:

$$I\ 127 + n^1_0 \rightarrow I\ 128 + \text{gamma}$$

$$C_5H_{12} + I\ 128 \xrightarrow{\text{during emission}} C_5H_{11}\ I\ 128 + H.$$

Processes of this type can, perhaps, be used for synthesizing chemical compounds containing active atoms, both for the purposes of chemical research and for the production of highly radioactive preparations with a definite physiological action.

The formation of radioactive iodine according to the reaction I 127 (n, gamma) I 128 does not depend on the valence state of the stable iodine exposed to the neutrons. According to Knauer's data [21], during exposure of iodine in the form I', IO'_3 and I', IO'_3, I_2 the change in formation of radioactive iodine is between 2 and 3%.

The effective cross-section (sigma) of the nuclear reaction

$$I\ 127 + n^1_0 \rightarrow I\ 128 + \text{gamma}$$

and its dependence on the energy of neutrons were studied by many authors [22-29]. For fast (Rn—Re) neutrons, V. S. Dementiy and D. V. Timoshchuk [29] determined that sigma equals $6.57 \cdot 10^{-24}$ cm^2. For slow neutrons the effective cross section was at least 100 times larger. Figure 3 shows the dependence, obtained by Jones [27], of the cross-section of I on the neutrons' energy in the range from 0.0026 eV to 1000 eV. For thermal neutrons (E equals 0.025 eV) the cross-section of reaction (n,gamma), taking into account the cross section of scattering, equals sigma sub-o equals $6.7 \cdot 10^{-24}$ cm^2. When E<3 eV, the interaction of the iodine with the neutrons follows the law 1/v; when E equals 20.3 eV, a resonance line occurs; and in the range E equals 25 —50 eV, there is a resonance band containing at least three resonance maxima.

Append

Fig. 3. Cross-section of I as a function of Neutron Energy

SECRET

A radioactive iodine of half-life T equals 25 minutes can be obtained not only by the nuclear reaction studied above, but also by other methods. Livingood and Seaborg [29] separated iodine of half-life T equals 25 ± 1 minute from tellurium subjected to bombardment with deuterons of energy E equals 8 MeV. In this case I 128 was formed from a stable isotope of Te 128 (32.8%) in the reaction Te $^{128}_{52}$ (d,2n) I $^{128}_{53}$ with discharge of 1 active atom per $2 \cdot 10^7$ deuterons with E equals 8 MeV. Formation of I 128 in this manner was corroborated by Tape [16], who also obtained it by bombarding lead iodide with 7 MeV deuterons, obviously, according to the reaction I $^{127}_{53}$ (d,p) I $^{128}_{53}$. Furthermore, this isotope of iodine is formed in the bombardment of tellurium with protons [9] in a (p,n) reaction.

Since the half-life was very exactly determined for I 128, this isotope can be used to calibrate instruments that measure radioactivity. According to the data of Hull and Seelig [30], I 128 has a half-life T equals 24.99 ± 0.02 minutes.

The radiation emitted by I 128 during beta-disintegration was studied by Fermi, Amaldi, and other authors [8], who, having separated I of half-life 25 minutes, demonstrated the ejection of electrons and gamma-quanta by this isotope and also determined the absorption coefficient of beta-particles in aluminum. The beta-spectrum of I 128 was first prepared by the Soviet scientists Alikhanov, Alikhanyan, and Dzhelepov [31, 32] with the aid of an original device with two coincidence counters in the magnetic field. The beta-spectrum thus obtained is shown in Figure 4. Here E_m equals 2.1 MeV. Most of the electrons have energy E equals 0.5 MeV. M. I. Korsunskiy, N. N. Nikolayevskaya, and M. A. Bak [7] studied the beta-spectrum of I 128 in the energy range from 0.6 to 2.0 MeV by the same method of magnetic analysis with two coincidence counters. Using a more powerful source, they made a more detailed study of the beta-spectrum of I of half-life 25 minutes in the above-mentioned energy range.

SECRET

SECRET

The beta-spectrum of I 128 was also studied in many laboratories in a variety of ways. The results of individual determinations were not always in good agreement. Livingood and Seaborg [8] determined the absorptive layer for the beta-rays of I 128 as equal to 1.05 g/m² Al, which when recalculated according to Feather's formula corresponded to E_m equals 2.23 MeV. In measuring, with a Wilson chamber and magnetic field, of the radii of curvature of 1330 tracks, Tape [16] obtained from certain products a beta-spectrum with E_m equals 2.4 MeV. He also gave [33] another value, E_m equals 2.2 MeV. Plotting the F. Curie graph, in coordinates $(\frac{N}{f})^{1/K}$ and E for K = 2 and K = 4, gives by extrapolating in the former case E_m = 2.2 MeV and gives in the latter case E_m = 2.94 MeV. This forces us to conclude that the beta-spectrum of I 128 follows the original theory of Fermi rather than the theory of Konopinskiy-Yulenbek.

On the other hand, Bacon, Grisewood, and van der Merwe [34, 35] also studied the spectrum of beta-particles emitted from I 128, with the aid of a Wilson chamber and magnetic field, and analyzed the Curie graph for K = 4. They started by proposing to apply the Konopinskiy-Yulenbek theory to the beta-spectrum of iodine of half-life 25 min. Moreover, on this assumption the discontinuity in the Konopinskiy-Yulenbek graph (Figure 5) was interpreted by the authors as an indication of the presence of a complex spectrum in I 128. By breaking the Konopinskiy-Yulenbek graph into two straight lines and extrapolating them up to the intersection with the axis of the abscissae, they obtained for the values of the limiting (boundary) energies of two groups of beta-particles: E_m = 1.05 MeV and E_m = 2.10 MeV. These data are set forth in the well-known tables of Seaborg and Maddox. But these values are open to doubt, both because they were obtained by the questionable application of Konopinskiy-Yulenbek's theory to the beta-decay of I 128 and because they do not agree with other experimental data.

Fig 4. Beta-Spectrum of I 128

SECRET

Fig 5. Graphs of Fermi and Konopinskiy-Yulenbek for the beta-spectrum of I 128 (according to Bacon, Grisewood, and Van der Merwe)

SECRET

In fact, according to the data of other authors [7, 16], the theory of Konopinskiy-Yulenbek is not in sufficiently good agreement with the experimental values of the maximum energy of the beta-spectrum of I 128. Incidentally, the authors [35] themselves do not offer any definite conclusions about the applicability of the Konopinskiy-Yulenbek theory. On the other hand, to solve the problem of the presence in I 128 of two groups of beta-particles of energy E_m = 1.05 MeV and E_m = 2.10 MeV, the character and energy of the gamma-rays emmitted by the Xe 128 nucleus, during the radioactive disintegration I 128 Xe 128, are of the greatest importance. It is obvious that, if the values obtained by Bacon, Griscwood, and van der Merwe are correct, fairly intensive gamma-rays with E_g = 1.05 MeV must be observable in the beta-decay.

But according to data from many laboratories it is known that even if I 128 emits gamma-rays in a beta-decay, [5, 14, 36], their intensity is very low and their energy is less than one MeV. Thus, Roberts and Irvine [37], and even Bacon, Griscwood, and van der Merwe [35] were not able to detect gamma-rays in a quantity greater than 1 photon per 10 beta-particles. Even if the energy of these photons corresponded to the difference between the limiting (boundary) energies of two groups of beta-particles, it would be very doubtful that such an insignificant quantity of beta-particles (10%), the emission of which was followed by the radiation of gamma-quantum, could have any important effect on the form of the beta-spectrum and the corresponding Curie graph.

Further, the absorption measurements of the energy of gamma-rays made by M. A. Bak and N. N. Nikolayevskaya and by Livingood and Seaborg give a more consonant value for the energy of gamma-rays of 25-minute iodine, which does not at all correspond to the above-mentioned values of the maximum values of the two groups of beta-particles.

A more satisfactory conception of the decay of I 128 was suggested by Siegbahn and Hole [39], who studied the beta-radiation of this isotope with the aid of a beta-spectrograph and determined the energy of gamma-rays by the energy of photoelectrons produced from lead.

SECRET

13

A preparation of radioactive iodine was placed in a copper capsule, with lead foil 0.1 millimeter thick in front of it. The beta-particles emitted by I 128 were completely absorbed in the walls of the capsule. The secondary electrons were studied with the beta-spectrograph. Figure 6 shows the spectrum of secondary electrons emerging during the action of the gamma-rays of the radioactive iodine on the copper capsule and lead foil. In the background of Compton electrons two peaks can be seen which correspond to photoelectrons from K and L levels of the lead layer. The energy of gamma-rays as computed from the energy of photoelectrons, taking into account the energy of the K and L levels in connection with Pb, is consonant and equal to E = 0.428 MeV. Gamma-rays of higher energy were not detected. These results are in agreement with the data obtained by absorption measurements. Gamma-rays of energy E = 0.428 MeV have a little intensity and, as shown by the pulse recordings in beta and gamma counters, are emitted at the rate of 7 gamma-quanta per 100 beta-particles. Thus, the remission of gamma-quanta accompanies only 7% of the beta-disintegrations and, consequently, the beta-spectrum of I 128 must consist of two groups with a difference between the maximum values of E_{max} equaling 0.428 MeV.

Fig 6. Spectrum of the secondary electrons formed by the action of the gamma-rays of I 128

Fig 7. Fermi's graph for the beta-spectrum of I 128

Fermi's graph in Figure 7 gives E equal to 2.02 MeV, which agrees well enough with the value obtained by Alikhanov, Alikhanyan, and Dzhelepov.

It is apparent that, because of the small intensity in the second group of beta-particles, their presence is not indicated in Fermi's graph. In accordance with the statements made above, the transformation I 128 → Xe 128 might be represented by the scheme in Figure 8 in agreement with Siegbahn and Hole. 92% of the beta-particles are emitted in transition to the basic level of the Xe 128 nucleus, 7% are emitted in the transition

SECRET

to the excitation level of Xe 128 with an energy intensity of 1.59 MeV and a subsequent radiation of gamma-quanta with Eg = 0.428 MeV.

Fig 8. Diagram of the transition $I^{128} \rightarrow Xe^{128}$.

~~I 129. T is about 10 years.~~

I 129. T is About 10^8 Years

For a long time no one succeeded in detecting the iodine isotope of mass number 129 either among the stable or radioactive isotopes.

In studying the isomerism of Te 129, Seaborg, Livingood, and Kennedy [40] discovered the isomeric transition of Te 129 (32 days) Te 129 (72 days). According to Nier [1], however, I 129 was not stable or more accurately, it was not contained in raw iodine in quantities larger than 1/40,000. Hence, I 129 either had to be a very rare stable isotope or it had to have a very long half-life.

Actually, it follows from the published data on the plutonium project [41] that I 129 is formed during fission of U 235, has a "very long" half-life and emits electrons during its transformation into stable Xe 129. I 129 is formed as a result of a chain of beta-transformations of primary fission-products emerging during the fission of uranium according to the following scheme:

$$Sb^{129}_{51}\ (4.2\ hrs) \longrightarrow \begin{cases} Te^{*129}_{52}\ (32\ days) \\ Te^{129}_{52}\ (72\ min) \longrightarrow I^{129}_{53}\ (very\ long) \longrightarrow Xe^{129}_{54}\ (stable) \end{cases}$$

In analyzing the integral decay curve of iodine isotopes separated from neutron-bombarded uranium, it was not possible to discover a half-life greater than 8 days. Even when the action upon uranium had continued for 123 days in a Clinton furnace [42], the iodine separated from it produced a decay curve containing only known half-lives and ~~disintegrated~~

SECRET

15

decayed to the limit T = 8 days which corresponds very well with I 131. But this specimen of iodine subjected, after decay to the threshold, to the action of slow neutrons, displays the considerable energy attributed to iodine of half-life T = 12.5 hours. From its half-life and absorption coefficient in Al this activity was identified as I 130. It formation must, obviously, be ascribed to radiation neutron-capture of an iodide of mass M = 129. The effective cross section of the reaction I 129 (n, gamma) I 130 can be roughly estimated as equal to sigma = $8 \cdot 10^{-24}$ cm 2.

The very long half-life I 129 (greater than 10^8 years) permits us to assume its presence in raw iodine in quantities approximately 80 times smaller than the limit set by Nier.

I 130. T = 12.6 hours.

Livingood and Seaborg (8) (45) isolated a radioactive isotope of half-life T = 12.6 hours, emitting electrons during decay from tellurium bombarded with deuterons of energy 8 MeV. This half-life was ascribed to I 130 formed from stable Te 130 (33.1%) according to the reactions $Te\ {}^{130}_{52}(d,2n)\ I\ {}^{130}_{53}$ with a discharge of 1 atom per 1.107 deuterons of energy E = 8 MeV. During bombardment of tellurium with deuterons of energy 14 MeV, the total discharge of I 130 is $32 \cdot 10^6$ fissions per micro-A per hour, which corresponds to the discharge of one active atom per 1.1×10^4 deuterons.

Some iodine isotopes are formed in bombarding tellurium with deuterons. Figure 9 shows the complex decay curve for iodine separated from deuteron-bombarded tellurium and its analysis (according to Livingood and Seaborg).

Fig 9. The Decay Curves of Radioactive Isotopes of Iodine formed during Bombardment of Te with Deuterons.

A convincing proof that the half-life T = 12.6 hours belongs to I 130 is furnished by the fact that iodine of half-life T = 12.6 hours is also obtained in bombarding cesium with fast neutrons (44). Since cesium has only one stable isotope Cs $^{133}_{55}$, the formation of radioiodine results from the reaction.

$$Cs\ ^{133}_{55}\ (n, \text{alpha})\ I\ ^{130}_{53}$$

But this isotope was obtained in exposing tellurium to the action of protons (9) in the obvious reaction: Te $^{130}_{52}$ (p,n) I $^{130}_{53}$.

Statements contained in the bibliography to the effect that radio-active iodine of half-life about T = 13 hours can be isolated from the fission products of uranium (45) and Thorium (45), (46) by the action of neutrons, should be accepted with care. The presence of some radioactive isotopes of iodine formed in these cases, the decay of certain isotopes with the formation of active products and the change in the relations between the intensities of discrete activities depending upon the conditions of exposure make it extremely difficult to analyze the integral decay curve. It is very probable that, under definite conditions of exposure, the 22-hour half-life of I 133 may be concealed by the presence of the 6.6 hour half-life of I 135 and the 9.4-hour half-life of Xe 135 springing from it. Obviously it is due to this circumstance that in various laboratories, in analyzing the integral decay curve of radioactive iodines isolated from the fission-products of heavy nuclei, a large number of intermediate periods were obtained with T equal to: 12 hours (45); 15--16 hours (46)(47); 18.5 hours (48).

Thus Polesitskiy and Orbeli discovered a period, T = 15--16 hours, which occurred during the fission of thorium, which may belong to both I 130 and I 133. This circumstance is noted by various authors (47).

SECRET

The presence of I 130 among the fission products of uranium has evidently been treated in the most recent works (41).

In the fission of U 235, stable Te 130 is formed as primary fission-product; hence, a chain of beta-transformations, leading to I 130, cannot appear.

The radiation emitted by I 130 beta-~~disintegration~~ decay was studied by means of its absorption (6) in Al and Pb in a Wilson ~~camera~~ chamber with a magnetic field (16) and, in greater detail, by Roberts, Downing, Elliott and Deutsch (11), (49), (50) using a magnetic spectrometer and the coincidence method.

Figure 10 shows the beta-spectrum of I 130 obtained by these authors. Four peaks can seen in it corresponding to internal conversion electrons. The energies of the corresponding gamma-quanta, computed while taking into account the energies of K-capture in Xe, equal: E_1 = 0.416 ± 0.005 MeV; E_2 = 0.538 ± 0.007 MeV; E_3 = 0.665 ± 0.008 MeV; E_4 = 0.747 ± 0.010 MeV. Approximate values are obtained for Eg by analyzing the spectrum of secondary electrons formed by the action of gamma-rays of I 130 on Au, Pb, Sn (0.417; 0.535; 0.670; 0.740 MeV).

p. 1 Fig 10 p. 1

Fig 10. Beta-Spectrum of I^{130}

In studying the diagram of I 130 decay it is very important fact that these 4 types of gamma quanta are not all equally likely. The intensity of the line of Eg = 0.416 MeV amounts only 0.3 ± 0.1 of the mean intensity of the other gamma-lines. Consequently, the gamma-quantum of energy Eg = 0.416 MeV is not emitted in every act of beta-~~disintegration~~ decay. On the other hand, study of the change in the number of beta-gamma coincidences, appearing per each recorded beta-particle in dependence upon the thickness

SECRET

18

of the absorbent placed between beta-counter and source, makes it possible to establish that, when the energy E of a beta-particle is greater than 0.6 MeV, the number of beta-gamma coincidences does not depend on the thickness of the absorbent. These experimental factors give rise to the conclusion that the beta-spectrum of I 130 must consist of two groups of beta-particles, one of which of energy E = 0.6 MeV, (approx.) is accompanied by the emission of 4 gamma-quanta, while the second is accompanied by the same gamma-quanta, but emits the quantum of energy Eg = 0.416 MeV. These conclusions agree with the data obtained by analyzing a Fermi graph plotted by the above-mentioned authors for the beta-spectrum obtained by them for I 130 (Figure 11). Using coordinates $(\frac{N}{f})$ and E, we obtained a broken line resolving into two straight lines for which give two values for the maximum energy of beta-particles: $E'_m = 0.61 \pm 0.02$ MeV and $E''_m = 1.03 \pm 0.02$ MeV. The value $E'_m = 0.61$ MeV is in good agreement with the results of research on beta-gamma coincidences, and $E''_m = 1.03$ MeV practically coincides with the value $E_m = 1.05$ MeV obtained by Livingood and Seaborg through absorption measurements. The difference $E''_m - E'_m = 0.42$ MeV agrees exactly with Eg = 0.416 MeV.

Fig 11. Fermi's Graph for the Beta-Spectrum of I^{130}.

Fig 12. Decay diagram of I^{130}, Xe^{130}

Thus the transformation of I 130 → Xe 130 must correspond in principle with the diagram given in Figure 12. Two groups of gamma-particles of energies $E'_m = 0.61$ MeV and $E''_m = 1.03$ MeV are emitted. In both cases there is a transition to the excitation level of Xe 130. Both transitions are accompanied by emission of gamma-quanta in cascade. As a result, the Xe 130 nucleus is converted into its primary state. The order of emission of gamma-quanta

except the gamma-quantum of energy Eg = 0.416 MeV has not been determined and is arbitrarily indicated in the diagram.

SECRET

I 131 . T = 8 days.

In bombarding tellurium with deuterons, Tape and Cork(14) obtained activity of half-life T equaling 8 days. This they wrongly attributed to Te 131. Soon afterwards Liningood and Seaborg (8), (15) demonstrated that this eight-day activity was chemically identical with iodine and was formed during bombardment of tellurium. Iodine of half-life 8 days was obtained directly by the reaction Te $^{130}_{52}$(d,n) I $^{131}_{53}$ and by the beta-disintegration of a radioactive isotope of Te 131 originating in the activation of tellurium by deuterons and neutrons:

Te 130 (d,p) Te 131; Te 130 (n,gamma) Te 131; Te $^{131}_{52}$ → I $^{131}_{53}$ + beta-minus

The appearance of I 131 directly in the process of the nuclear reaction Te $^{130}_{52}$(d,n) I $^{131}_{53}$ proceeds with the following rates: 1 atom per 5.10^8 deuterons (8) when the energy of the deuteron is 8 MeV; 1 atom per 0.7.10^4 deuterons (43) when E = 14 MeV.

As two isomeric states are known for Te 131 with half-lives T = 30 hours and T = 25 minutes, it is interesting to ascertain which of the isomers is the direct predecessor of I 131 when the latter is formed during the beta-disintegration of Te 131. As shown by Seaborg and Kennedy (40), (51), I 131 is formed from the 25-minute isomer of Te 131, whereupon the following chain of transformation reactions takes place:

Te $^{131}_{52}$ —30 hrs.→ Te $^{131}_{52}$ —25 min.→ I $^{131}_{53}$ —8 days→ Xe $^{131}_{54}$ (stable).

The existance of just such a mechanicism for the formation of I 131 in the beta-disintegration of Te 131 was proved through the chemical method of separating isomers (52) applied if the isomers were genetically connected (if one isomeric state arose from another in isomeric transformations). The idea of this method is that, as a result of the emission experienced by the nucleus in greatly transformed isomeric gamma-transistions, a change occurs in the valence state of the atom undergoing isomeric transition. As a result, atoms in the process of isomeric transition and in their

SECRET

SECRET

primary state can be separated from atoms in a mestastable state.

It seems that, if to deuteron-exposed telluric acid there is added inactive tellurous acid and if for some time So_2 is bubbled through a 3N solution of HCl to reduce elementary Te, then the precipitate will contain only tellurium of half-life 25 minutes, which breaks down with the formation of iodine of half-life 8 days.

In connection with the problem of the isomeric transmutations of Te 131, preceding the formation of I 131, the unproved but undisputed statement of Roberts and Irvine (10) is curious. They separated from tellurium, activated by deuterons of energy 11.5 MeV repeatedly during the course of several months, a radioactive iodine which differed very little from 8-day I 131 as to half-life according to absorption measurements. If this activity is identical with I 131, it will become necessary to assume the existence of a third long-lived isomer in Te 131.

The formation of I 131 takes place every time Te 131 appears as a result of any process whatever. This isotope of iodine was observed by Abelson (53),(54),(55) and by Hahn and Strassman (48),(56) in fission products of neutron-bombarded uranium. Fermi and Segrè (57) separated it from uranium exposed to alpha-particles of energy 32 MeV.

I 131 is not the primary fission-product of uranium, but originates as the result of beta-decay of Te 131, whereupon the following chain occurs:

SECRET

$$Te\ {}^{131}_{52} \xrightarrow{30\ hours.} Te\ {}^{131}_{52} \xrightarrow{25\ min} T\ {}^{131}_{53} \xrightarrow{8\ days} Xe\ {}^{131}_{54}\ (stable).$$

The yield of this chain during uranium fission was studied by many authors. Proceeding from the work of Jentschtke (58),(59), it is possible to assume that there should be no great difference in the coefficients yield in these two cases. In fact, the yield coefficients for a chain of mass number 131 during uranium fission are very close for slow (41),(60),(61) and fast (61) neutrons and amount to 2.2---2.8% of the total number of fissions. Yaffe and Mackintosh (62) determined the ratios of the activities I 131 and Ba 140 which he separated from uranium by exposing it to slow

21

SECRET

22

and fast neutrons, and obtained an almost identical value for these coefficients. According to the values previously obtained for yields of Ba^{140} in uranium fission for slow and fast neutrons[62] the yield coefficients for I^{115} were determined as equalling, respectively: 2.25±0.11% and 2.27±0.11%.

Radiation and Plot of Decay Curve. The beta-spectrum of eight-day iodine was studied by its absorption in Al[8],[61],[61] with the help of a Wilson chamber and magnetic field[16] and of a magnetic spectrometer[63],[64]. The most trustworthy of these varied data are those of Deutsch, Dawning and Roberts. With the help of a magnetic spectrometer and research work on beta-gamma coincidences, the latter discovered that I^{131} had a simple spectrum with E_m = 0.595±0.01 MeV. The maximum value of the beta-spectrum, E_m = 0.595 MeV, was obtained Both from the beta-spectrum itself (Figure 13) and from the corresponding graph of Fermi (Figure 13) and from the corresponding graph of Fermi (Figure 14). This value agrees with the value E_m = 0.6 MeV obtained from absorption measurements[41].

~~-171-~~

Figure 13. Beta-spectrum of I^{131}.

The energy of the gamma-rays of I^{131} were determined by the absorption in Pb[8],[11] and by the energy of secondary[16],[49],[64] and conversion[49],[63],[64] electrons, whereupon the results of measurement by different methods were in sufficiently good agreement. According to Downing, Deutsch and Roberts[50],[64] each beta-particle was accompanied by the emission of two gamma-quanta in cascade with energy E_g = 0.357±0.007 MeV and E_g = 0.080±0.001 MeV. The order of the emission of gamma-quanta was not determined.

/171/

SECRET

Figure 14. Fermi's graph for the beta-spectrum of I^{131}.

Figure 15. Diagram of the transformation $I^{131} \rightarrow Xe^{131}$.

A probable diagram of the transformation $I^{131} \rightarrow Xe^{131}$ is shown in Figure 15. It consists of the emission of beta-particles of energy E_m = 0.595 MeV followed by transition to the excitation level of Xe^{131} and of the subsequent radiation of two gamma-quanta in cascade followed by transition to the primary state of Xe^{131}.

$I^{(132)}$. T = 2.4 hours

Beta-activity of half-life T = 2.5 hours was first discovered by Meitner, Hahn and Strassmann[65] in neutron-bombarded uranium. It was also established that it did not belong to the primary product of uranium and neutron interaction, but occurred as the result of a series of beta-disintegrations. In accordance with the ideas current at the time in regard to "transuraniums", this 2.5-hour activity was attributed to EkaPt, originating according to the following nuclear reaction:

$$U + n \rightarrow U_{92} \xrightarrow{10\ \text{secs}} EkaRe_{93} \xrightarrow{2.2\ \text{min}} EkaOs_{94} \xrightarrow{59\ \text{min}} EkaIr_{95} \rightarrow$$

$$\rightarrow EkaPt_{96} \xrightarrow{2.5\ \text{hrs}} EkaAu_{97}.$$

Soon after the discovery of uranium fission, Adelson[66] and Feather and Bretscher[67] demonstrated almost simultaneously that the 2.5-hour activity was chemically identical with iodine and that its predecessor of half-life about three days (66--77 hours) was tellurium. These results were obtained by analyzing the absorption of a series of radiation products emitted by "transuranium" elements and by chemical isolation of activities with the corresponding carriers. They provided a new and independent proof of the fission of uranium nuclei.

SECRET

SECRET

It seems strange that the typical metalloid iodine could have been identified previously as EkaPt. It is interesting that weight proofs were brought forward on this point. 66-9 and 2.5-hour activities were separated out from an acid solution of neutron-irradiated uranium salt by means of hydrogen sulfide with platinum. Then the 2.5 hour activity was separated by crystallizing ammonium chloroplatinite. In the light of the ideas of that time as to "transuraniums", all this seemed good and sufficient proof to the chemical identity of 2.5 hour activity with EkaPt. As a matter of fact, precipitation of Te occurred which, upon beta-disintigration, gave 2.5-hour iodine and adsorption of I in surface-active sulfur precipitates; also, joint crystallization occurred with the ammonium chloroplatinite of the corresponding iodine group.

The chain of beta-~~disintegrations~~ decays arising from the primary fission-products obtained during uranium fission by neutron bombardment and leading to radioactive iodine was studied in detail by Axelson (53)(54), Academician V.G. Khlopin with his fellow- workers (68),(69),(70),(71) and by Hahn and Strassmann (3),(48),(56). Finally, all the elements of the chain were chemically identified and their half-lives were accurately defined. Sb was the primary fission-product according to the following transformation:

$$\underset{51}{Sb} \xrightarrow{\sim 5\ \text{min}} \underset{52}{Te} \xrightarrow{77\ \text{hrs}} \underset{53}{I} \xrightarrow{2.4\ \text{hrs.}} \underset{54}{Xe}\ (\text{stable})$$

The iodine isotope under consideration is formed not only during uranium fission but, as shown by Hahn, Strassmann and Fluegge (72) and by Polesitskiy and Nemorovskiy (73), also during thorium fission by the action of neutrons. It is also separated from uranium subjected to bombardment by alpha-particles of energy 32 MeV (57).

Since, as far as we know, radioactive iodine of half-life T = 2.4 hours was separated only from fission products of heavy nuclei and was not obtained in other nuclear reactions, all its characteristics cannot be indicated with sufficient definiteness. This refers, in the first place, to the mass number of the 2.4 hour isotope. It seems likely that it should be

SECRET

attributed to the radioactive iodide of mass number 132, but it is difficult to prove this hypothesis.

The second characteristic--the half-life--can be determined accurately enough, in spite of the complicated nature of the integral curve of radio-active isotopes of iodine separated from the fission products of heavy nuclei. This is possible because of a fission-product of 2.4 hour iodine has a long half-life T = 77 hours, whereas all the other radioactive isotopes of iodine are formed from short-lived isotopes of tellurium. Consequently 2.4-hour activity can be separated practically in its pure form in secondary extraction of iodine from radiated salt. By rational selection of the time for the first and second extraction of iodine, it is possible to obtain a sufficient quantity of pure I 132 and to determine its first half-life without resorting to analysis of the integral curve with its attendant errors. From a series of such determinations we obtain T = 2.3 --2.4 hours.

The yield coefficient of 2.4-hour iodine in uranium fission by the action of neutrons amounts to 3.3--3.6% (41),(61). The radiation emitted by 2.4 hour iodine has not been sufficiently studied. The data in the bibliography are not numerous and do not agree among themselves. According to the absorption measurements of Born and Seelmann-Eggerbert(74) this isotope of iodine has a simple beta-spectrum with E_m = 1.35 MeV, and emits gamma-rays with Eg = 0.85 MeV. According to data brought forward in a published account of the plutonium project (41) and also according to data obtained through absorption in Al and Pb, the beta-spectrum of 2.4 hour I 132 consists of two groups with Em = 1.0 MeV (50%) and Em = 2.1 MeV (50%), and the emission of two gamma-quanta of energy Eg = 0.60 MeV (50%) and Eg = 1.4 MeV (50%).

Such contradictory results do not allow of plotting a decay scheme for this isotope of iodine.

I 133. T = 22 hours.

The radioactive isotope of iodine of half-life T = 22 hours was first discovered by Adelson(53), (64), in fission products of neutron-bombarded uranium.

This was confirmed by Hahn and Strassmann (48),(56) by V.G. Khlopin with his fellow-workers (69), by Dodson and Fowler (75) and by others. This radio-active iodine was also obtained in uranium fission by the action of alpha-particles (57) and, obviously, in the exposure of thorium to the action of fast neutrons (45),(47),(76),(77). The study of this isotope presents definite difficulties. Since 22-hour iodine was obtained only in the fission of heavy nuclei, it was always studied in a mixture with the other radioactive isotopes of iodine formed in this case and with their active decay products. In this case it was not possible to separate the activity under consideration in the pure form successfully isolated in the case of I 132. Under such conditions the fact of the separation of 22-hour activity with inactive iodine does not prove that this activity belongs to iodine. Special experiments must be conducted to offer convincing proof that this activity really belongs to iodine and not to the decay product of some isotope of iodine (48). Determination of the half-life is only possible by analyzing the complex decay curve displaying the decomposition of several iodine isotopes with different yield coefficients, the half-lives, the energies of beta-particles, and the complicated nature of the change in their decay products. (Figure 16). As the nature of the integral curve is determined by many factors, among them the conditions of target exposure, the character of the counter arrangement used, and so forth, the difficulties arising in the determination of the half-life are obvious. Out of the many determinations (42),(48),(53),(55),(69),(77) the most trustworthy value seems to be T - 22 hours.

Fig 16. Example of an Integral Decay Curve for Radioactive Isotopes of Iodine. a represents 8-day I^{131}; b, 22 hour I^{133}

In spite of these difficulties, not only was the period of 22-hour iodine established but it was shown by successive fissions that this radioactive iodine was not a primary fission-product but was formed from tellurium of half-life T - 60 minutes, which, in turn, resulted upon beta-decay of Sb of half-life T less than 10 minutes (55). Hence, this chain occurs:

$$Sb_{51} \xrightarrow{10\ min} Te_{52} \xrightarrow{60\ min} I_{53} \xrightarrow{22\ hours} Xe_{54}.$$

But this chain does not end in xenon. Many authors (71), (78), (79) had already remarked on the fact that radioactive xenon arises from iodine separated from the fission products of uranium and thorium. By successive separation of xenon from the iodine isolated from uranium, Segrè and Wu (77), (80) and Dodson and Fowler (75) showed that, in the disintegration of 22-hour iodine, radioactive xenon of half-life of about 5 days was formed, which, in turn, decayed to form stable cesium.

This circumstance was utilized by Wu (44), (80) in determining the mass number of I and of the whole chain. Cesium, with one stable isotope Cs 133, was subjected to neutron bombardment. A radioactive gas was then obtained which, through its half-life and the absorption curve of the radiation emitted by it, was identified with 5-day Xe separated from 22-hour iodine. Since, obviously, the nuclear reaction is $Cs\ 133_{54}$, the mass number 133 must be attributed to 22-hour iodine and all its fission products.

On the basis of the above statements the following chain takes place:

$$Sb^{133}_{51} \xrightarrow{10\ min} Te^{133}_{52} \xrightarrow{60\ min} I^{133}_{53} \xrightarrow{22\ hrs.} Xe^{133}_{54} \xrightarrow{5.4\ days} Cs^{133}_{55}\ (stable).$$

The yield of this chain during the fission of U 235 equals 4.5% (41).

The radiation emitted by I 133 in beta-decay, was studied with the aid of a Wilson chamber and magnetic field (81) and by its absorption in Al and Pb (41). The values obtained by this method for the maximum energy of the beta-spectrum of I 133 are equal, respectively to: Em = 1.4 MeV and Em = 1.3 MeV. The energy of the gamma-rays according to absorption measurements equals Eg = 0.55 MeV. In spite of the fairly satisfactory coincidence of values for Em obtained by two different methods of evaluating the reliability of data on the beta-spectrum of I 133, it is difficult to

separate it from other radioactive isotopes. It is necessary to proceed with all proper precautions. In particular, the method employed to obtain the specimen of I^{133} in Perfilov's[81] experiments, in principle, tolerates an error in the determination of the beta-spectrum of I^{133}. On the basis of data on the yield coefficients of chains leading to the formation of radioactive isotopes of iodine and on the half-lives of isotopes forming part of these chains[41], it is easy to show that considerable quantities of other isotopes of iodine ($I^{131,135}$) and xenon (Xe^{133}, Xe^{135}) must inevitably be present during the course of irradiation in the experiment and the subsequent extraction of radio-iodine separated from uranium before its introduction into a Wilson ~~camera~~ chamber, mixed with I^{133}. Even ~~in~~ the fairly arbitrary assumption that every xenon atom formed in AgI crystals—in the time interval between the separation of iodine from uranium and the beginning of the measurements—is completely ~~scattered~~ diffused in the atmosphere; the specimen contains I^{133} which is far from pure. Incidentally, even the nature of a Fermi graph, plotted for the beta-spectrum obtained[81], indicates that ~~even one~~ not a single group of beta-particles occurs.

I^{134} .T = 54 minutes.

Like the previous isotope of iodine, the 54-minute I^{134} was separated only from the fission-products of heavy nuclei. This isotope was first discovered by Adelson[53,54,55] in analyzing the decay curve of radio-iodine separated from uranium exposed to the action of neutrons. By this method 54-minute iodine was next obtained in many laboratories[48,56,60].

But this isotope was also obtained in the fission of uranium by the action of alpha-particles[57] and, as shown by Dodson and Fowler[45] and Polesitskiy and Orbeli[46,47], could be separated from thorium exposed to the action of neutrons. 54-minute iodine was formed during the separation of heavy nuclei, but not in the form of a primary fission-product. It is a product of a series of beta-disintegrations. The chain, including 54-minute iodine, was studied by a number of authors[46,47,53,54,55,70] and is now represented in the form: $Sb_{52}^{134} \xrightarrow{10\ min} Te_{52}^{134} \xrightarrow{43\ min} I_{53}^{134} \xrightarrow{54\ min} Xe_{54}^{134}$ (stable). The half-lives of the elements forming part of the chain and preceding the 54-minute I, were established by studying the changes in the activity of iodine (consequently of tellurium) in successive emissions from tellurium separated from uranium (or uranium solution).

There are the same difficulties in studying I^{134} as in studying I^{133}.

The yield coefficient of 54-minute I in the fission of uranium by slow neutrons is approximately equal to 5.7%[41].

The mass number of 54-minute iodine was determined very recently. In Seaborg's[9] table of isotopes, 54-minute I was called I^{131} and in the report on the plutonium project[41] it was hypothetically given the mass number 134 ($I^{(134)}$).

Proof of this hypothesis by neutron bombardment with atomic numbers close to iodine, with protons, duetrons, and alpha-particles was not possible. There are no suitable stable isotopes in Sb, Te, and Cs and active Xe can give no clear-cut answer. However, there is another way of solving the

problem. In 1947, in determining the relation of the activities of 54-minute I and Ba^{139}, in accordance with the known yield of Ba^{139} during the fission of uranium[62], Yaffe and Mackintosh[61] obtained a yield coefficient for 54-minute I in fission both by slow and fast neutrons equal, on an average, to 5.75%. On the other hand, it is known from the mass-spectrographic measurements of Thode and Graham that in the fission of uranium four stable isotopes of xenon are formed: Xe^{131}; Xe^{132}; Xe^{134}; Xe^{136} which are the final products of a chain with corresponding mass numbers. The ratios of yield of the isotopes of xenon with mass numbers 132,134, 136 to Xe^{131} and consequently the ratios of the yield coefficients of the corresponding chains equal: k_{132}/k_{131} = 1.48 - 1.50; k_{134}/k_{131} = 2.63 - 2.67; k_{136}/k_{131} = 2.17 - 2.30.

k_{131} was determined for 8-day I^{131} according to the ratio of the activities of I^{131} and Ba^{140} and equaled 2.23 - 2.27%[61].

Hence it follows that the yield coefficient of a chain with M = 134 equals 5.85% which, within limits of error, corresponds with the yield of 54-minute iodine. Such a method, by the way, may be used to determine the mass number of 2.4-hour iodine.

The yields of Ba^{139} and Ba^{140}, the starting-points in determining the yield coefficients of 54-minute I and 8-day I were determined for fission by slow neutrons in accordance with the ratio of the activities of U^{239} and Ba^{139} (consequently of Ba^{140}) from the ratio

$$\frac{\text{beta-activity of } U^{239}}{\text{beta-activity of } Ba^{139}} = \frac{\text{sigma sub-c}}{\text{sigma sub-f} \cdot k_{139}},$$

where sigma sub-c is the cross section of the capture of U^{238}

and sigma sub-f is the cross section of the fission of U^{235}; k_{139} is the yield coefficient of Ba^{139} in the fission of U^{235} by fast neutrons (the ratio of the number of fissions leading to the formation of Ba^{139} to the total number of fissions). The yields of Ba^{139} and Ba^{140} in fission by fast neutrons are determined by the ratio of the corresponding activities in the fission of U^{235} and U^{238}.

The radiation of I^{134} has scarcely been studied. It is merely known that in the beta-~~disintegration~~ decay of I^{134} gamma-rays are radiation with E_g greater than 1 MeV.

I^{135} .T = 6.6 hours.

Segrè and Wu[77],[80] discovered still another isotope of iodine with the half-life T = 6.6 hours in the fission products of heavy nuclei. This radio-active iodine, the presence of which was confirmed by other authors[47],[60],[75], was formed during the decay of short-lived tellurium[75],[77], originating directly in the fission of uranium and thorium.

Xenon formed in the beta-~~disintegration~~ decay of I^{235} is radioactive,[75],[77], which made it possible to establish both the presence of 6.6-hour iodine among the fission products of heavy nuclei and also its mass number.

It is difficult directly to determine the existence of a 6.6-hour half-life by the integral decay curve for radio-active iodine separated from neutron-irradiated uranium and thorium. The great number of activities composing this curve conceals the 6.6 hour half-life and for this reason it has been determined successfully only in rare instances. In general, analysis of the integral curve has only succeeded in recording the fact of

the growth of an activity belonging to a radioactive descendant of 6.6-hour iodine, xenon, with a half-life of 9.2--9.4 hours. In practice the half-life of I^{135} was established by a series of successive emissions of xenon from iodine separated from neutron-irradiated uranium and thorium.

The separation of xenon from iodine is brought about either by blowing off by gas through a boiling alkaline solution of sodium iodide or by the method of Langsdorf and Segrè[79] which consists of preparing an emanation specimen from which xenon passes into an exhaustion chamber. The specimen is prepared in the following manner. Silica gel is saturated with silver nitrate by immersion for 15 minutes in a 0.1N solution of $AgNO_3$ and then dried for some hours at a temperature of 70°C. Several cubic centimeters of silica gel prepared in this manner are shaken up with a solution of active iodine in CCl_4 until the solution becomes discolored. The active xenon formed in the emanation sample passes either into the exhaustion chamber surrounding the counter or directly inside the counter.

The xenon separated in one way or the other from the iodine, contains two isotopes: Xe^{133} of half-life T = 5.4 days and Xe^{135} of half-life T = 9.4 hours. However, because of the great difference in the periods and the insignificant energy of the electrons emitted by Xe^{133} (Xe^{133} - E_m ~ 0.3 MeV; Xe^{135} - E_m ~ 0.95 MeV) under the conditions of the experiment, the xenon separated from the iodine actually disintegrates in a half-life[80] T = 9.4 hours. In Figure 17, points on the semilogarithmic coordinates represent the decay

of xenon for four extractions made at 12-hour intervals. The curve of the "progenitor" of 9.4-hour Xe radio-iodine of half-life T = 6.6 hours, was obtained in extrapolating the decay curves (straight lines in the coordinates, Figure 17) to the moment of separation of xenon from iodine along the initial points denoted by circles. Starting from the fact that 9.4-hour Xe is formed during the beta-~~disintegration~~ decay of 6.6-hour I and that, consequently, they both have the same mass number, it was possible to prove that the period of 6.6 hours belonged to an iodine isotope of mass number 135. In bombarding Ba with neutrons[44,80] two radioactive gases were obtained with half-lives T = 5 days and T = 9.4 hours, which according to their half-lives and absorption curves were identified with radioactive isotopes of xenon formed during the decay of iodine separated from the fission products of uranium and thorium. Although barium has 7 stable isotopes, the obvious nuclear reaction Ba_{56}^{A} (n, alpha) Xe_{54}^{A-3} leads to radio-xenon only for the mass numbers 130, 136, and 138. Since Xe^{133}, which, as proven above, has T≈5 days, is formed from Ba^{136}, and the ratio $Ba^{130}:Ba^{138}$ = 1/700, it is possible to draw the conclusion that 9.4-hour Xe is formed in the nuclear reaction: Ba_{56}^{138}(n, alpha) Xe_{54}^{135}. Consequently, the 6.6-hour half-life belongs to I^{135}.

/178/

Figure 17. The decay curve of I^{135}.

With regard to the decay scheme of I^{135}, it has been established that Xe^{135}, originating in the beta-decay of 6.6-hour iodine is formed in two isomeric states[80,83,84] of Xe^{*135} with T = 10---13 minutes and Xe^{135} with T = 9.2--9.4 hours.

Goette[83], who studied radioactive xenon formed from iodine with the aid of a Seelmann-Eggebert[85] apparatus, was the first to discover 10-minute Xe. He showed that the half-lives T = 10 minutes and T = 9.4 hours did not belong to discrete isotopes of xenon formed from various isotopes of iodine, but that both had as their "progenitor" 6.6-hour I and, consequently, belonged to two isomers of one isotope of Xe^{135}.

Active iodine is separated from neutron-irradiated uranium by the application of the usual carrier technique. Radioactive xenon is carried away from a heated solution of sodium iodide by a flow of hydrogen and is absorbed by active carbon at the temperature of liquid air. In a subsequent heating of the carbon the "dissorbed" xenon passes into an exhaustion chamber surrounding a Geiger-Mueller counter. The hydrogen is blown off at two-hour intervals. The decay curve of the radio-isotopes of xenon can be located each time on two straight lines with T = 10 minutes and T = 9.4 hours. The activity with T = 5 days belonging to Xe^{133} was not observed, evidently, both because of the short period of accumulation and because the beta-particles of Xe^{133} were absorbed to a considerable extent by the walls of the counter. Extrapolating both straight lines with T = 10 minutes and T = 9.4 hours to the initial moment of time, Goette obtained for the initial

activities in both cases a straight line with exactly the same inclination.

By separating xenon from iodine at short intervals of accumulation and analyzing the decay curves obtained by recording the beta-particles and gamma-quanta, Wu and Serge[80] discovered that T = 10 minutes belonged to the excited isomeric state of Xe^{135}. This isomer was obtained in the beta-decay of I^{135} to the amount of about 10% and with emission of gamma-quanta (and internal conversion electrons), it passed into the basic state of Xe^{135}, decaying with T = 9.4 hours.

According to recent data[41], the transmutation chain of the primary fission-products with M = 135, including the formation and decay of I^{135} can be represented in the following form:

$$Te_{52}^{135} \xrightarrow{<2\ min} I_{53}^{135}\ (6.7\ hrs) \xrightarrow{\sim 10\%} Xe_{54}^{*135}\ (10\ min) \xrightarrow{} Xe_{54}^{135} \xrightarrow{9.4\ hrs} Cs_{55}^{135} \xrightarrow{2.5\times10^{6}\ yrs} Ba_{56}^{135}\ (stable).$$

The yield of this chain in uranium fission by thermal neutrons is equal to 5.9%.

The energy conversions which take place during the beta-decay of I^{135} are not accurately established. According to the data on hand obtained by the absorption method[41], the maximum energy of beta-particles emitted by I^{135} lies between E_m = 1.35--1.5 MeV and the energy of the gamma-quanta radiated during beta-decay lies between E_γ = 1.6--1.3 MeV.

The difference between the energy levels of the basic and metastable states of Xe^{135} equals about 0.55 MeV[41],80,84.

The half-life T = 1.8 minutes was provisionally attributed[41] to I^{136} formed as a primary fission-product in the fission of uranium and decomposing into stable Xe^{136}.

If it is true that the 1.8 minute half-life belongs to I^{136} and that the following process takes place:

$$I_{53}^{136} \xrightarrow{1.8\ \text{min}} Xe_{54}^{136}\ (\text{stable}),$$

the yield of 1.8 minute I during uranium fission must equal 4.85%.[61]

Seelmann-Eggebert and Born[84,87] demonstrated that 30-second iodine decomposed to form Xe^{137} with half-life T = 3.4 minutes. According to American data[41] 30-second I^{137} is formed directly during fission and decomposes according to the scheme:

$$I_{53}^{137} \xrightarrow{30\ \text{secs}} Xe_{54}^{137} \xrightarrow{3.4\ \text{min}} Cs_{55}^{137} \xrightarrow{\text{ce yrs}} Ba_{56}^{137}\ (\text{stable}).$$

According to these same data[41,88], in uranium fission one more short-lived isotope of iodine is formed as a primary fission-product with T = 22--23 seconds. It decomposes with the ejection of electrons and delayed neutrons.

Within the range of accuracy of the experimental data on hand, the decay scheme of I^{135}, starting from the general rules of "radiation" of excited nuclei, can be hypothetically represented in the following form: in beta-~~disintegration~~ decay 100% of the beta-particles are emitted with energy E_m = 1.35--1.5 MeV, whereupon a transition occurs to the excited level of Xe^{135} with an energy of excitement E_{exc} = 1.6--1.3 MeV. This excited state with emission of gamma-quanta "radiates" to basic or metastable level of Xe^{135}. Approximately 90% of the gamma-quanta, following the emission of beta-particles, have an energy E_g = 1.6--1.3 MeV and hence in 90% of all cases a transition takes place to the basic state. 10% of the gamma-quanta have an energy $E_g = E_{exc}$--0.55 $\approx$ 0.9 MeV and in this case there is a transition to the metastable state of Xe135 which, with the half-life T = 13 minutes, is discharged through conversion gamma-transitions with energy E = 0.55 MeV. According to absorption measurements the group of gamma-quanta with energy Eg $\sim$ 0.9 MeV may be detected as a result of the slight degree of intensity (10%).

Short-lived isotopes of iodine:

$I^{(136)}$. T = 1.8 min; I^{137} . T = 30 sec; $I^{(137)}$. T = 22 sec.

Strassmann and Hahn[86] succeeded in working out a method of separating radio-active iodine from neutron-irradiated uranium such that measurements of the activity of AgI* could begin within 3 minutes after the end of the exposure. Thereby they discovered in the fission-products of uranium two short-lived isotopes of iodine with half-lives T = 1.8 $\pm$ 0.4 minutes and T = 30 $\pm$ 6 seconds.

Tables I and II give the characteristic constants of radioactive isotopes of iodine and the nuclear reactions leading to the formation of these isotopes

/181/

Table I

M	Type of Radiation	T	E_m in Mev	E_γ in Mev	Reactions in Formation
124		4 days			
(125)	K-capture	56 days			
126		13 days			
128		24.99 minutes			
129		$\geq 10^8$ years			
130		12.6 hours			
131		8 days			
(132)		2.4 hours			
133		22 hours			
134		54 minutes			
135		6.6 hours			
(136)		1.8 minutes			
137		30 seconds			
(137)		22 seconds			

[Translator's note: See Table for figure spacing]

NOTE

SECRET

/182/

Table II

Radioactive Isotopes of Iodine, Formed in the Fission of Uranium[4]

M		Coeff. of Yield Discharge in Fission of U^{235} in %
129	Sb (4.2 hrs) → Te* (32 days) / Te → 72 min → J → $\geq 10^8$ yrs → Xe	0.7
131	Te (30 hrs) → Te → 25 min → J → 8 days → Xe	2.8
(132)	Sb → ~5 min → Te → 77 hrs → J → 2.4 hrs → Xe	3.6
133	Sb → < 10 min → Te → 60 min → J → 22 hrs → Xe → 5.4 days → Cs	4.5
134	Sb → < 10 min → Te → 43 min → J → 54 min → Xe	5.7
135	Te → < 2 min → J (6.7 hrs) → Xe (10 min) / J → 9.4 hrs → Xe → Cs → $\geq 2.5 \cdot 10^4$ yrs → Ba	5.9
136	J → 1.8 min → Xe	4.8
137	J → 30 sec → Xe → 3.4 min → Cs → 33 yrs → Ba	--

Conclusion

In nuclear reactions leading to the formation of radioactive isotopes of iodine, there is usually a simultaneous formation of several isotopes. This especially concerns the case of the fission of heavy nuclei. More than half the isotopes of iodine are formed by this process, and we do not know how to obtain many of them by any other methods. The fis-

SECRET SECRET

CONCLUSION

In nuclear reactions leading to the formation of radio-active isotopes of iodine, there is usually a simultaneous formation of several isotopes. This especially concerns the case of fission of heavy nuclei. More than halfthe isotopes of iodine are formed by this process, and we do not know how to obtain many of them by any other methods. The fission reaction of heavy nuclei, as a source of radio-iodine, has many advantages

SECRET

Pick Up Next Page

SECRET

and defects. On the one hand, it is the most accessible means of obtaining radioactive isotopes of iodine with extremely interesting half-lives. This method is accessible for any laboratory possessing (Ra--Be) or (Rn--Be) neutron sources, and does not require any complicated high-voltage apparatus. Iodine obtained by this method consists of a number of isotopes with half-lives ranging from a few seconds to several days. This makes it possible to solve many problems. On the other hand, this same simultaneous formation of a number of isotopes in which it is difficult to distinguish one from the other, makes the method somewhat defective. This defect can be overcome to a certain degree by varying the time of exposure, extraction of the exposed uranium, and extraction of the separated iodine. On the basis of the data in Table II, it is possible to make a rational selection of conditions for obtaining radio-iodine such that the optimum number of interesting isotopes may be obtained. It is also possible to separate the half-lives by utilizing a different energy of beta-particles and by employing corresponding filters in measuring activities.

The fact of the formation of radioactive isotopes of xenon in the decay of I^{133} and I^{135}, which causes alterations in the decay curve, is of great importance. In using the radio-iodine (active) containing these isotopes as an indicator, it is obviously necessary to take proper measures so that all the specimens of radio-iodine (active) involved in the various processes during the course of the experiment may remain under equal conditions in relation to the possibility of a diffusion of radio-xenon (active) from the specimen.

iodine involved in the various processes during the course of the experiment may remain under equal conditions in relation to the possibility of diffusion of radioxenon from the specimen.

Bibliography

1. Nier, A.O., Phys. Rev., 52, 933, 1937
2. Lu, C.C.; Sugden, S.; J. Chem. Soc., 1273, 1939
3. Hahn, O.; Strassmann, F., Naturwiss. 27, 451, 1939
4. Szilard, L.; Chalmers, T.A., Nature, 134, 462, 1934
5. Amaldi, E.; D'Agostino, O.; Fermi, E.; Pontecorvo, B.; Rasetti, F.; Segrè, E., Proc. Roy.Soc., A 149, 522, 1935
6. Erbacher, O.; Philipp, K., Zeits. Phys. Chemie, A 176, 169, 1936
7. Korsunskiy, M.I.; Nikolayevskaya, N.N.; Bak, M.A., ZhETF 9, 524, 1939
8. Livingood, J.J.; Seaborg, G.T., Phys.Rev. 54, 775, 1938
9. Seaborg, G.T., Rev. Mod. Phys., 16, 1. 1944
10. Roberts, J.; Irvine, J., Phys. Rev. 59, 936, 1941
11. Roberts, A.; Elliott, L.; Dawning, J.; Peacock, W.; Deutsch, M.; Phys. Rev. 64, 268, 1943
12. Reid, A.F.; Keston, A.S., Phys. Rev. 70, 987, 1946
13. Glendenin, L.E.; Edwards, R.R., Phys. Rev. 71, 742, 1947
14. Tape, G.F.; Cork, J.M., Phys. Rev. 53, 676, 1938
15. Livingood, J.J.; Seaborg, G.T., Phys. Rev. 53, 1015, 1938
16. Tape, G.F., Phys. Rev. 56, 965, 1939
17. Feather, N., Proc. Cambr. Phil. Soc. 34, 599, 1938

18. Kurie, F.; Richardson, J.; Paxton, H., Phys. Rev. 49, 368, 1936

19. Fermi, E.; Amaldi, E.; D'Agostino, O.; Rasetti, F.; Segrè, E., Proc. Roy. Soc. A 146, 483, 1934

20. Reid, A.F.; Phys. Rev., 69, 530, 1946

21. Knauer, F., Zeits. f. Physik, 120, 103, 1942

22. Lampinte, C.; Rasetti, F.; Phys. Rev., 58, 554, 1940

23. Rasetti, F., Phys. Rev. 58, 869, 1940

24. Sinma, K.; Yamasaki, F., Phys. Rev. 59, 402, 1941

25. Houtermans, F., Zeits. f. Physik, 118, 424, 1941

26. Wu, C.S.; Reinwater, L.J.; Havens, W.W., Phys. Rev. 71, 175, 1947

27. Jones, W.B., Phys. Rev. 72, 362, 1947

28. Griffiths, J.H.E., Proc. Roy. Soc, A 170, 513, 1939

29. Dementiy, V.S.; Timoshchuk, D.V., DAN 27, 926, 1940

30. Hull, D.; Seelig, H., Phys. Rev. 60, 553, 1941

31. Alikhanov, A.I.; Alikhanyan, A.I.; Dzhelepov, V.S., Nature 135, 393, 1935

32. Alikhanov, A.I.; Alikhanyan, A.I.; Dzhelepov, V.S., Phys. Zeits. Sowjetun. 10, 78, 1936

33. Tape, G.E., Phys. Rev. 55, 1135, 1939

34. Bacon, R.; Grisewood, E.; van der Merwe, C., Phys. Rev. 54, 315, 1938

35. Bacon, R.; Grisewood, E.; van der Merwe, C., Phys. Rev. 59, 531, 1941

36. Amaldi, E., Phys. Zeits. 38, 692, 1937

37. Roberts, A.; Irvine, J., Phys. Rev. 53, 609, 1938

38. Bak, M.A.; Nikolayevskaya, N.N., DAN 22, 316, 1939

39. Siegbahn, K.; Hole, N., Phys. Rev. 70, 133, 1946

40. Seaborg, G.T.; Livingood, J.J.; Kennedy, J.W., Phys. Rev. 57, 363, 1940

41. "Nuclei Formed in Fission", J. Am. Chem. Soc. 68, 2411, 1946

42. Katcoff, S., Phys. Rev. 71, 826, 1947

43. Clarke, E.T.; Irvine, J.W., Phys. Rev. 70, 893, 1946

44. Wu, C.S., Phys. Rev. 58, 926, 1940

45. Dodson, R.; Fowler, R, Phys. Rev. 55, 880, 1939

46. Polesitskiy, A.Ye.; Orbeli, M.L., DAN 28, 216, 1940

47. Polesitskiy, A.Ye.; Nemorovskiy, N.; Orbeli, M.L.; Baranchik, N.M., Izv. AN SSSR, Phys. Series, 5, 603, 1941

48. Hahn, O.; Strassmann, F., Naturwiss. 27, 529, 1939

49. Deutsch, M.; Roberts, A., Phys. Rev. 60, 362, 1941

50. Downing, J.; Deutsch, M.; Roberts, A., Phys. Rev. 61, 389, 1942

51. Seaborg, G.T.; Kennedy, J.W., Phys. Rev. 55, 410, 1939

52. Segré, E.; Halford, R.S.; Seaborg, G.T., Phys. Rev. 55, 321, 1939

53. Abelson, P., Phys. Rev. 55, 670, 1939

54. Abelson, P., Phys. Rev. 55, 876, 1939

55. Abelson, P., Phys. Rev. 56, 1, 1939

56. Hahn, O.; Strassmann, F., Phys. Zeits. 40, 673, 1939

57. Fermi, E.; Segré, E., Phys. Rev. 59, 680, 1941

58. Jentschke, W.; Pranki, F., Zeits. f. Physik. 119, 696, 1942

59. Jentschke, W., Zeits. f. Physik. 120, 165, 1943

60. Andersen, H.; Fermi, E.; Grosse, A., Phys. Rev. 59, 52, 1941

61. Yaffe, L.; Mackintosh, C., Canad. J. Res. B25, 371, 1947

62. Grummitt, W.; Guéron, J.; Wilkinson, G.; Yaffe, L., Canad. J. Res. B 25, 364, 1947

63. Deutsch, M., Phys. Rev. 59, 940, 1941

64. Downing, J.; Deutsch, M.; Roberts, A., Phys. Rev. 61, 686, 1942

65. Meitner, L.; Hahn, O.; Strassmann, F., Zeits. f. Physik. 106, 249, 1937

66. Abelson, P., Phys. Rev. 55, 418, 1939

67. Feather, N.; Bretscher, E., Nature, 143, 516, 1939

68. Khlopin, V.G.; Pasvik-Khlopina, M.A.; Volkov, N.F., DAN. 24, 117, 1939

69. Khlopin, V.G.; Pasvik-Khlopina, M.A.; Volkov, N.F., DAN 24, 847, 1939

70. Khlopin, V.G.; Pasvik-Khlopina, M.A., Volkov, N.F., DAN 24, 851, 1939

71. Khlopin, V.G., Izv. AN, Phys. Series, 4, 305, 1940

72. Hahn, O.; Strassmann, F.; Fluegge, S., Naturwiss. 27, 544, 1939

73. Polesitskiy, A.Ye.; Nemerovskiy, N.N., DAN 28, 218, 1940

74. Born, H.J.; Seelmann-Eggebert, W., Naturwiss. 31, 201, 1943

75. Dodson, R.; Fowler, R., Phys. Rev. 57, 966, 1940

76. Langsdorf, A., Phys. Rev. 56, 205, 1939

77. Segré, E.; Wu, C., Phys. Rev. 57, 552, 1940

78. Khlopin, V.G.; Pasvik-Khlopina, M.A.; Volkov, N.F., DAN 24, 665, 1939

79. Langsdorf, A..; Segré, E., Phys. Rev. 57, 105, 1940

80. Wu, C.S.; Segré, E., Phys. Rev. 67, 142, 1945

81. Perfilov, N.A.; DAN 33, 491, 1941

82. Thode, H.G.; Graham, R.L., Canad. J. Res. A 25, 1, 1947

83. Goette, H., Naturwiss. 28, 449, 1940

84. Seelmann-Eggebert, W., Naturwiss. 31, 491, 1943

85. Seelmann-Eggebert, W., Naturwiss. 28, 451, 1940

86. Strassmann, F.; Hahn, O., Naturwiss. 28, 817, 1940

87. Seelmann-Eggebert, W., Born, H.J., Naturwiss. 31, 59, 1943

88. Snell, A.H.; Levinger, J.; Wilkinson; Meiners and Sampson, Phys. Rev. 70, 111, 1946